Effects of Aerosols on Tropospheric Composition and Ozone Budget

An Interactive Three-dimensional Global Particle and Chemical Model (MOZART-PT)

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Brief Description of the Models

(1) MOZART (Chemical/transport model)

25 levels from surface to 40 km

2.8 degrees in latitude and 2.8 degrees in longitude

50 chemical species (N_2O, CH_4, H_2O, NO_y, HNO_3, N_2O_5, Cl_x, O_x, CO, C_2H_6, C_2H_4, C_3H_6, C_4H_{10}, ISO, PAN,MPAN.......)

(2) Sulfate aerosol particle model

surface emission

chemical reactions (gas and aqueous phase)

wet and dry depositions

Heterogeneous reactions on Sulfate aerosol particles

(3) Carbon aerosol particle model

surface emission

transformation from hydrophobic to hydrophilic

wet and dry depositions

Heterogeneous reactions on carbon aerosol particles

(4) Interactions between chemistry and particles (MOZART-PT)

Carbon model **<=>** ***MOZART*** **<=>** ***Sulfate model***

The Heterogeneous Reactions

(1) N_2O_5 + (sulfate) -> HNO_3

Reduction in NOx, and decrease in O_3

Recommend reaction coefficient by JPL (Υ =0.1)

(2) (OH, HO_2) +(sulfate) -> Products

Reduction in OH, and decrease in O_3

Reduction in H_2O_2 and decrease in formation of SO_4 (sulfate particles)

Recommend reaction coefficient by JPL (Υ >0.2)

(3) HNO_3 +(Black carbon) ->NOx

Increases in NOx and increase in O_3

Reduction in HNO_3 and decrease in ratio of HNO_3/NOx

Recommend reaction coefficient by JPL (Υ =0.03-0.05)

(4) O_3 + (Black carbon) -> Products

reduction in O_3

Recommend reaction coefficient by JPL (Υ =0.0002-0.0033)

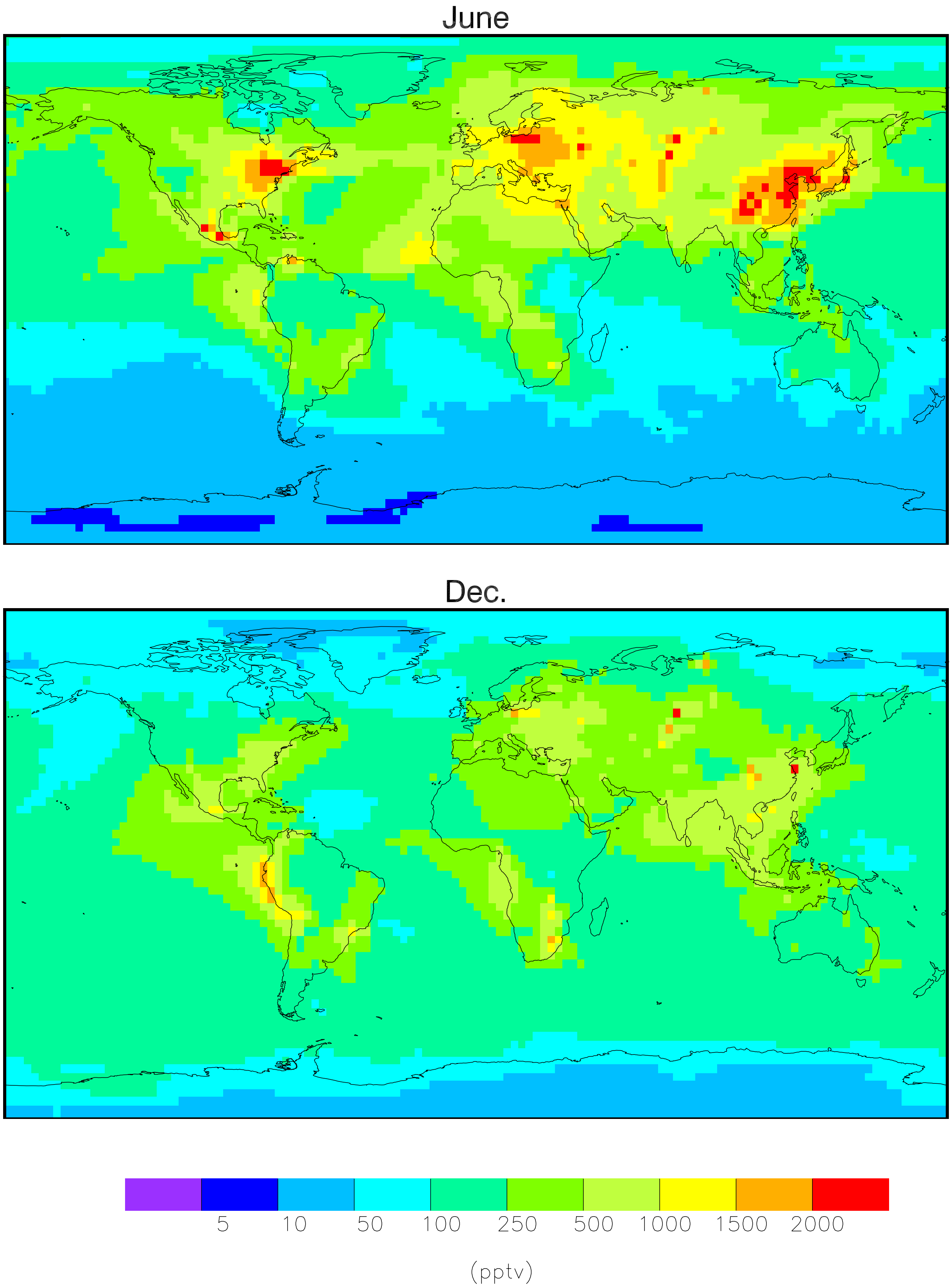
Fig. 1. Modeled surface distributions of SO_4 (pptv) in June (upper panel) and in December (lower panel).

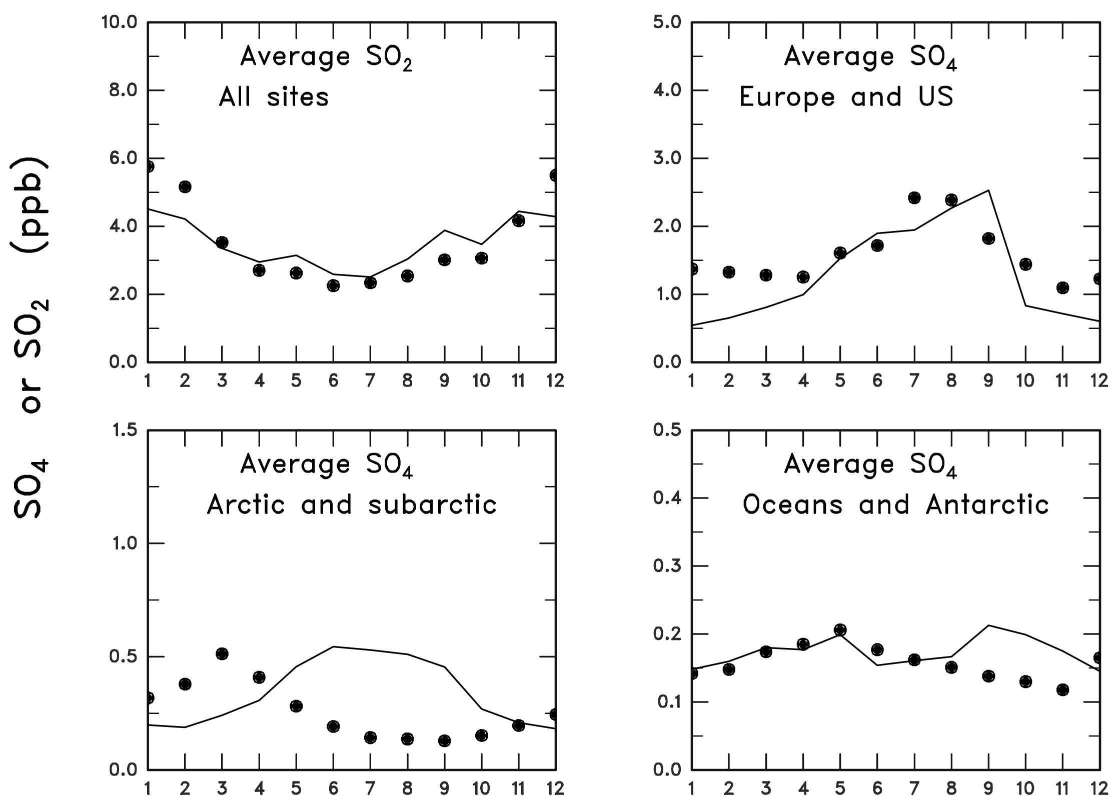
Fig. 2. Comparison of modeled with observed seasonal variations of SO_2 and SO_4 in different regions (Solid lines represent modeled results and dots represent observed results).

Fig. 3. Comparison of modeled with observed vertical profiles of SO_2 and SO_4 in North America (Solid lines represent modeled results and dots represent observed results).

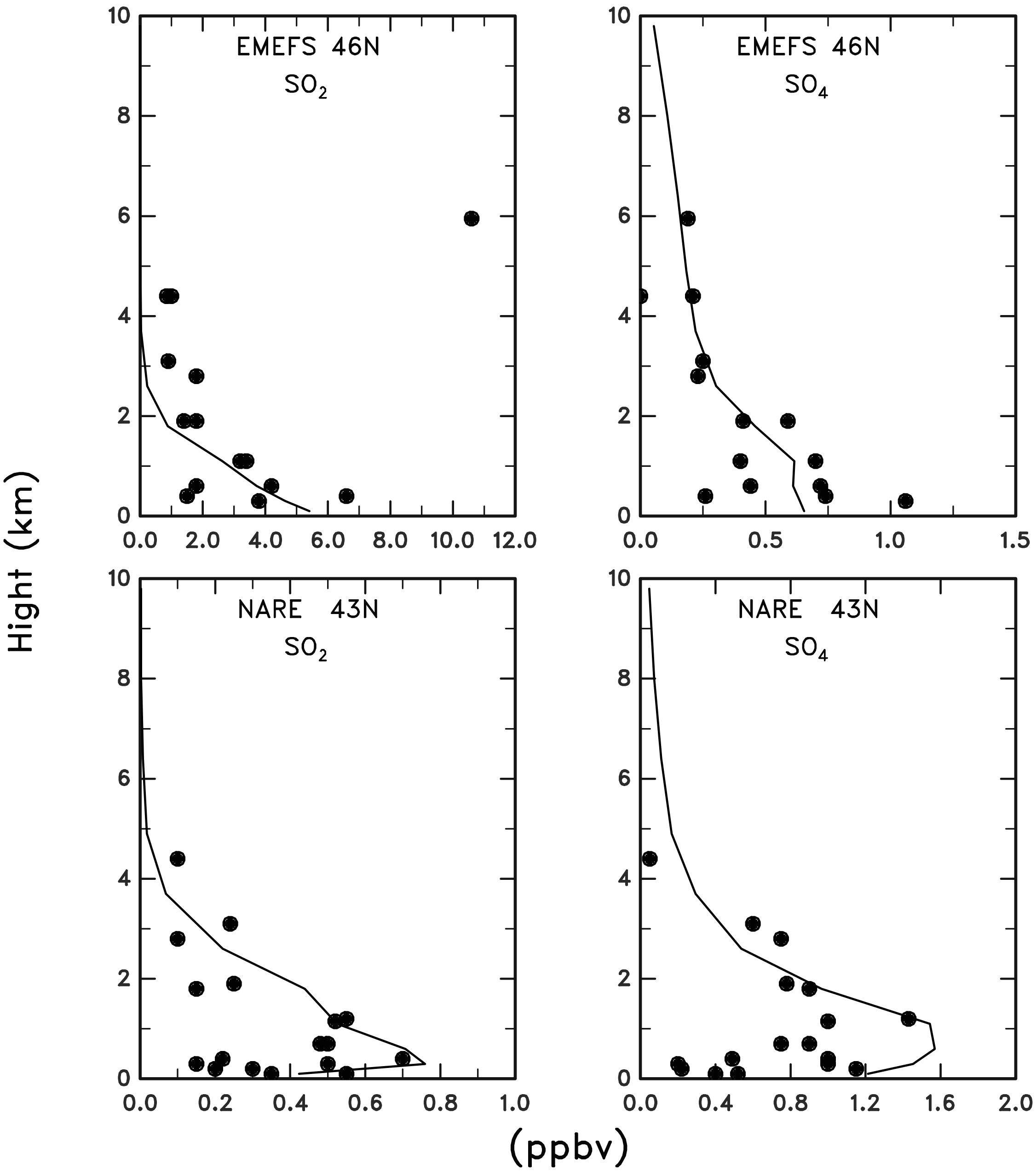
Fig. 4. Calculated changes (percent) in NO_x and O_3 as the consequence of the heterogeneous reaction of N_2O_5 + (sulfate).

Surface SO4

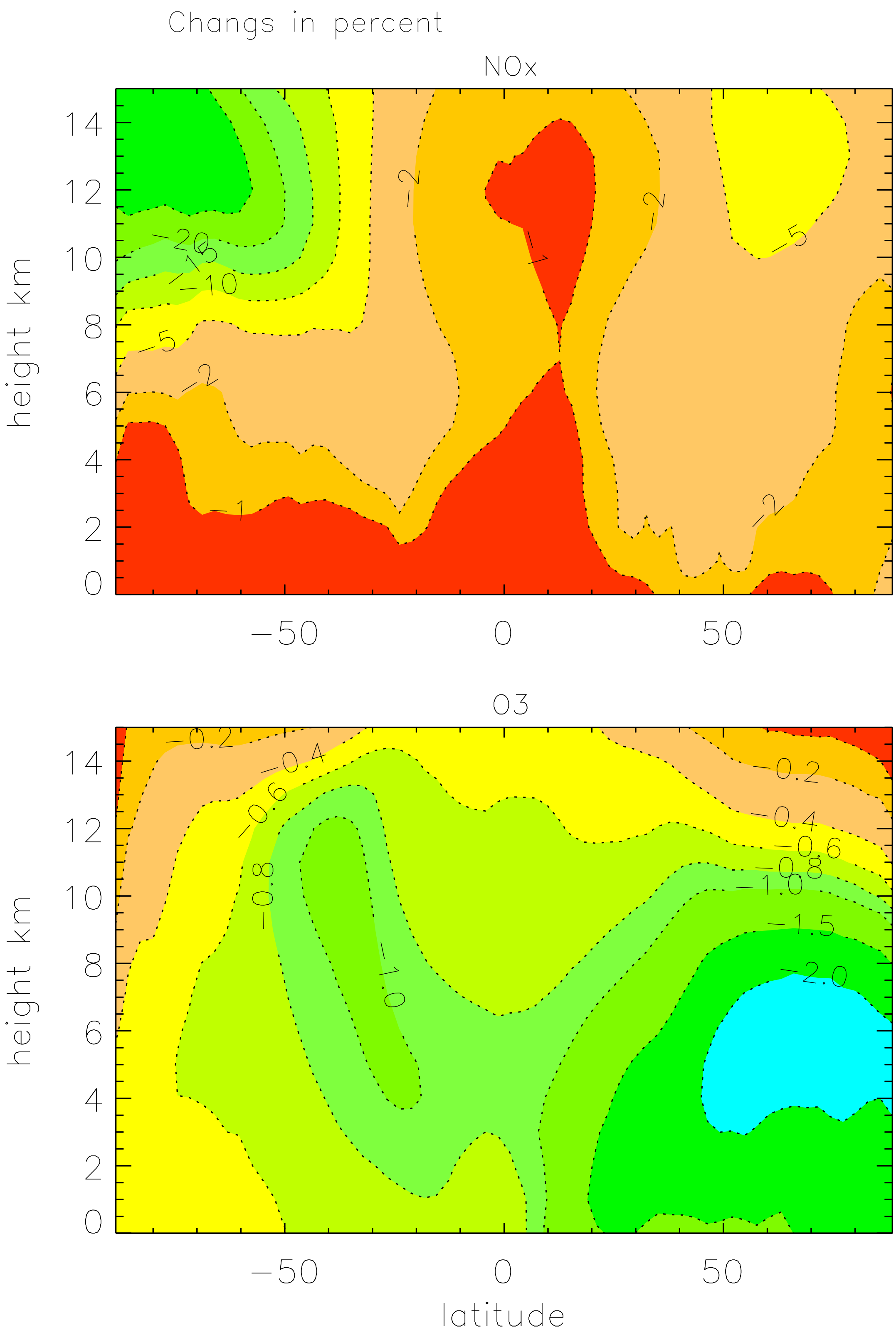




SO4 or SO2 (ppb)
Average SO2
All sites
Average SO4
Europe and US
Average SO4
Arctic and subarctic
Average SO4
Oceans and Antarctic



EMEFS 46N
SO2
EMEFS 46N
SO4
NARE 43N
SO2
NARE 43N
SO4
Hight (km)
(ppbv)



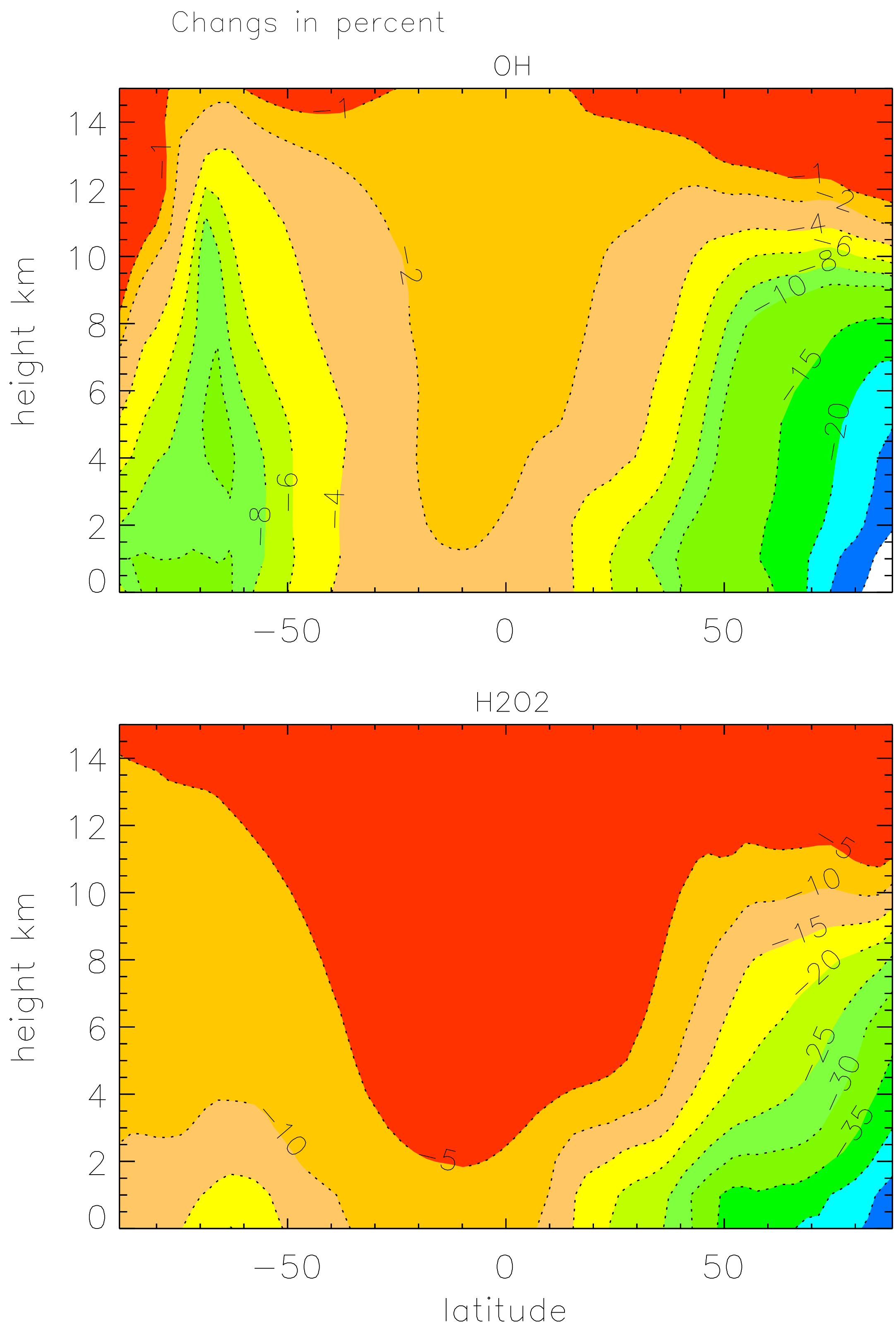
Changs in percent
NOx
height km
14
12
10
8
6
4
2
0
−50
0
50
−20
−15
−10
−5
−2
−1
−2
−1
−2
−5
−2
O3
height km
−0.2
−0.4
−0.6
−0.8
−1.0
−0.2
−0.4
−0.6
−0.8
−1.0
−1.5
−2.0
latitude

Fig. 5. Calculated changes (percent) in OH and H_2O_2 as the consequence of the heterogeneous reaction of HO_2 + (sulfate).

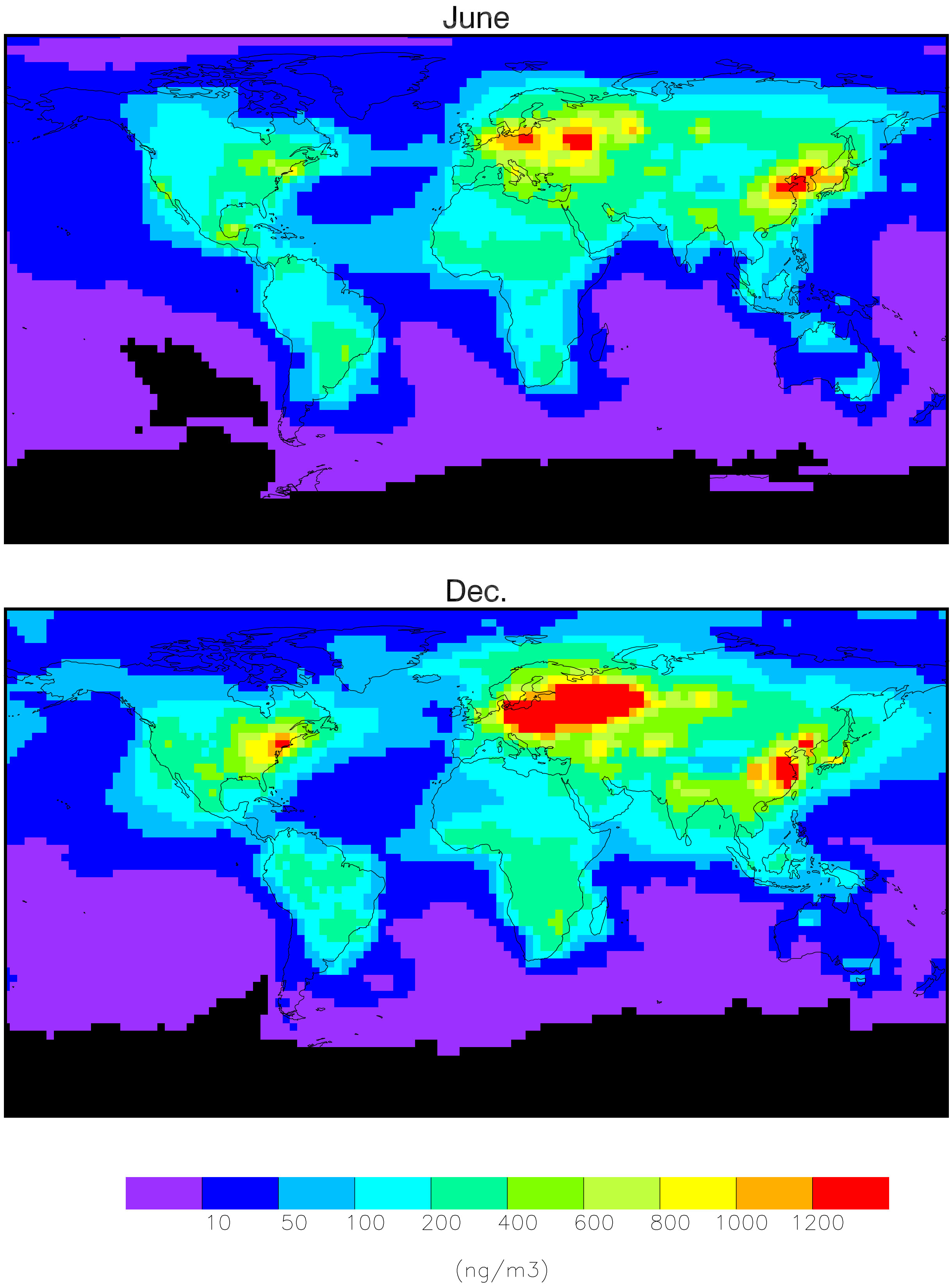
Fig. 6. Modeled surface distributions of black carbon (ng/m^3) in June (upper panel) and in December (lower panel).

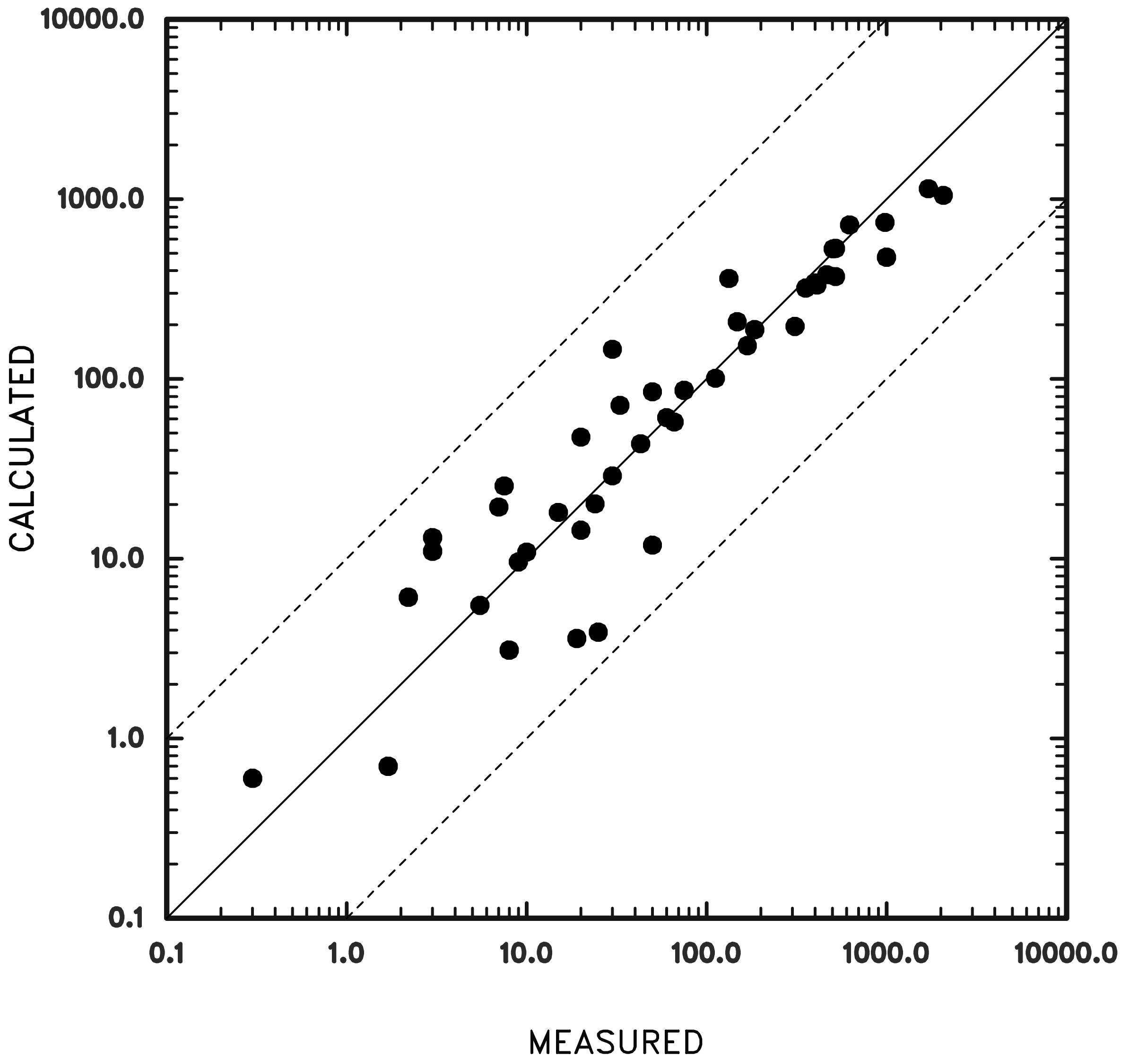
Fig. 7. Comparison of modeled with observed surface concentrations of black carbon (ng/m^3) (data is assembled by Liousse et al. 1996).

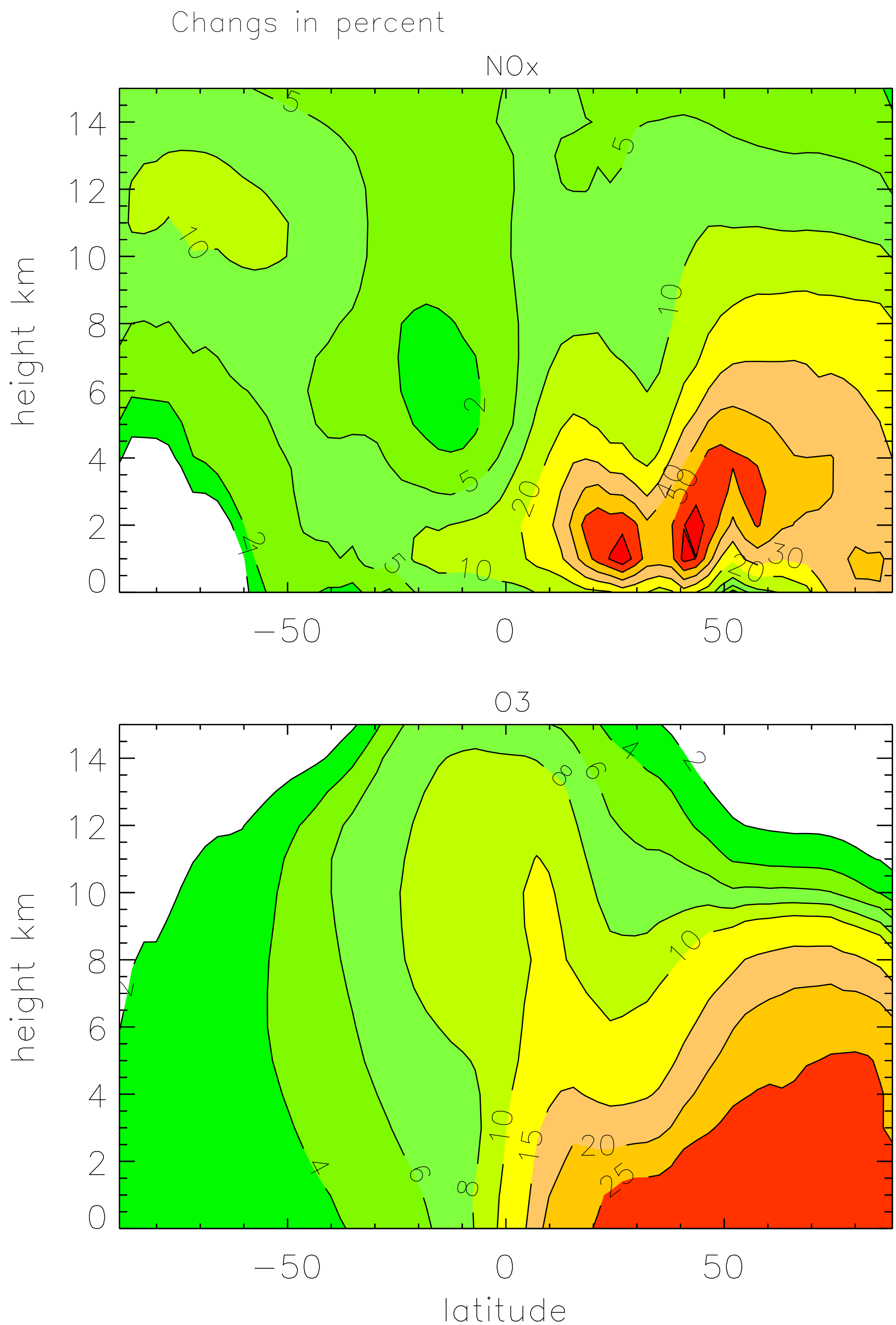
Fig. 8. Calculated changes (percent) in NO_x and O_3 as the consequence of the heterogeneous reaction of HNO_3 + (BC) -> NO_x.


Changs in percent
OH
H2O2
height km
latitude
−50
0
50
0
2
4
6
8
10
12
14
−1
−2
−4
−6
−8
−10
−15
−20
−5
−25
−30
−35

Surface BC

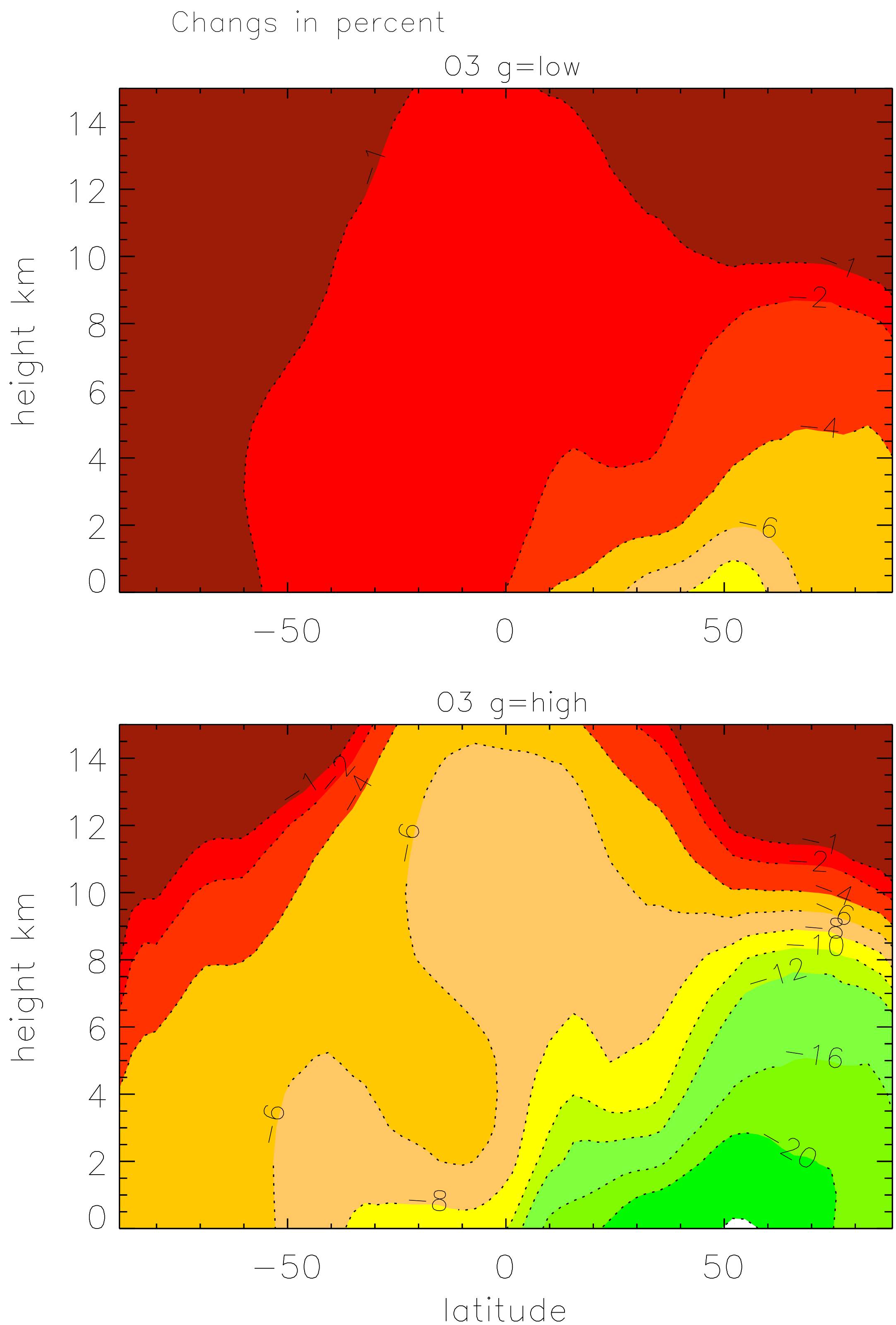



10000.0
1000.0
100.0
10.0
1.0
0.1
CALCULATED
0.1
1.0
10.0
100.0
1000.0
10000.0
MEASURED



Changs in percent
NOx
height km
O3
height km
latitude

Fig. 9. Calculated changes (percent) in O_3 as the consequence of the heterogeneous reaction of O_3 + (BC) by a low uptake coefficient (upper panel) and by a high uptake coefficient (lower panel).



Changs in percent
O3 g=low
O3 g=high
height km
latitude
14
12
10
8
6
4
2
0
−50
0
50
−1
−2
−4
−6
−8
−10
−12
−16
−20

Summaries

(1) The modeled global sulfate distributions are fairly consistent to observed global surface distributions (data is assembled by Chin et al. 1996). The seasonal variability in the Arctic region, however, is not successfully simulated. The comparison of vertical profiles between the model and observations (the airbone data is provided by Lohamann et al. 1998) is satisfactory. The modeled global carbon distributions are fairly consistent to observed global black carbon distributions (data is assembled by Liousse et al., 1996).

(2) There is a strong interaction between aerosol particles and chemical compounds in the troposphere. Aerosol particles affect the gas phase chemical species (e.g. H_2O_2), and the change in gas phase concentrations affects the formation of aerosol particles.

(3) Heterogeneous reactions on tropospheric aerosols (such as sulfate and black carbon aerosols) have the potential to play an important role in determining the chemical composition and the ozone budget in the troposphere. For example, the possible conversion of HNO_3 into NO_x improved the modeled HNO_3/NO_x ratio, and increased the concentration of NO_x near the surface, producing an O_3 increase by approximately 10 to 20 percent.